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Effects of Operating Variables on PAH Emissions and Mutagenicity of Emissions from Woodstoves

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Abstract

Since 1985, the U.S. Environmental Protection Agency (EPA) has conducted field emission measurement programs in Raleigh, NC, and Boise, ID, as part of the Integrated Air Cancer Project (IACP). These field studies were designed to identify the potential mutagenic impact of residential wood burning on ambient and indoor air. One facet of IACP involved obtaining emission samples from chimneys serving wood burning appliances. As a companion to the field source sampling, parallel projects were undertaken in instrumented woodstove test laboratories to quantify woodstove emissions during operations typical of in-house usage.

Three woodstoves were operated in test laboratories over a range of burnrates, burning eastern oak, southern yellow pine, or western white pine. Two conventional stoves were tested at an altitude of 90 m. One of the conventional stoves and a catalytic stove were tested at an altitude of 825 m. For the Raleigh support study, tests were started after a fire had been established and encompassed several wood additions over a 6-8 hour period. The Boise support tests were started with kindling a fire in a cold stove and continued for about 8 hours, including several wood additions. For the Raleigh support tests, emissions were collected using a modified EPA Method 5 sampling train. The woodstove dilution sampling system (WSDSS) was used for the Boise support tests.

As anticipated, the results showed wide variability, a common problem with woodstove testing. Total particulate emissions showed the expected inverse correlation with burnrate for the conventional stoves and nearly flat for the catalytic stove. While there appeared to be little or no correlation of total

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16. ABSTRACT The paper discusses studies in instrumented woodstove test laboratories to quantify woodstove emissions during operations typical of in-house usage. (The studies parallel field source sampling to identify the potential mutagenic impact of residential od burning on ambient and indoor air.) Three woodstoves were operated over a range of burnrates, burning eastern oak, southern yellow pine, or western white pine. Two conventional stoves were tested at an altitude of 90 m. One of the conventional stoves and a catalytic stove were tested at an altitude of 825 m. For one study, tests were started after a fire had been established and encompassed several wood additions over a 6-8 hr period. The other tests were started with kindling a fire in a cold stove and continued for about 8 hours, including several wood additions. For one test, emissions were collected using a modified EPA Method 5 sampling train. For the other tests, the woodstove dilution sampling system (WSDSS) was used. As anticipated, results showed wide variability, a common problem with woodstove testing. Total particulate emissions showed the expected inverse correlation with burnrate for the conventional stoves and nearly flat for the catalytic stove. While there seemed to be little or no correlation of total particulate emissions with altitude, the sur of the PAHs quantified showed an inverse correlation with altitude.

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particulate emissions with altitude, the sum of the specific polynuclear aromatic hydrocarbons (PAHs) quantified showed an inverse correlation with altitude: higher PAH emissions at the lower altitude.

Introduction

Since 1985, the U.S. Environmental Protection Agency (EPA) has conducted field emission measurement programs in Raleigh, NC, and Boise, ID, as part of the Integrated Air Cancer Project (IACP). These field studies were designed to identify the potential mutagenic impact of residential wood burning on ambient and indoor air. One facet of IACP involved obtaining emission samples from chimneys serving wood burning appliances. As a companion to the field source sampling, parallel projects were undertaken in instrumented woodstove test laboratories to quantify woodstove emissions during operations typical of in-house usage. The results from these laboratory source tests are the subject of this paper.

Experimental Design

Nearly all of the woodstove data in the literature have been obtained in a laboratory setting with the start of an emission test occurring after the fire was lit and the appliance brought up to operating temperature. In many cases, these tests also used dimension lumber as fuel as specified in various regulatory procedures[1,2,3]. In not-too-severe winter climates such as found in Raleigh, NC, and Boise, ID, it is common practice for woodstove users to kindle a new fire in a cold stove in the morning. This fire is often allowed to die out during the day when heating demand falls. A new fire is kindled during the early evening which is then stoked for the night and, often times, burns out before the residents rise the next morning. But a substantial percentage of users maintain a fire overnight, especially in colder climates. Since the objective of this work was to obtain emission samples under operating conditions similar to those observed in the field study areas, it was decided that some emission tests would start with lighting a fire in a cold stove (cold start), and others with a fire already established (hot start). Fresh wood was added several times during all tests. The experimental procedures followed have been described in detail elsewhere[4-7].

To limit the number of tests required to obtain statistically valid results, the number of operating variables to be investigated was limited to four in each test series. Each of these variables was investigated at two levels. For the Raleigh support tests, the variables investigated were:

Fuel----------eastern oak and southern yellow pine
Fuel moisture--seasoned (ave. 17%) and green (ave. 33%)
Fuel load------large (ave. 15.2 kg) and small (ave. 8 kg)
Burnrate-------high (ave. 6.6 kg/hr) and low (ave. 1.8 kg/hr)

A Sears, Roebuck and Co. catalogue number 42 G 84156N stove, manufactured by Atlanta Stove Works, Inc. (Model number 4040) was used for the Raleigh support

tests.

For the Boise support tests, the four variables investigated were:

Fuel----------eastern oak and western white pine
Burnrate------high (ave. 4.5 kg/hr) and low (ave. 2.3 kg/hr)
Stove type----conventional (Scott) and catalytic (Earth Stove 1000C)
Altitude------90 and 825 m

To prepare for an emission test, the stove and flue pipe were brushed and vacuumed clean. For cold start tests, a pretest fire was lit and burned for several hours at the conditions of wood species and burnrate planned for the next test. The pretest fire was allowed to burn out and the stove to cool to room temperature. For cold start tests, several newspaper balls and kindling wood were placed in the stove on the ashes from the pretest fire. All sampling equipment was started when the paper was ignited. The stove loading door was left open for 5 - 10 minutes until a good fire was established. At this time additional wood was loaded into the stove and the door closed. Each emission test lasted for about 8 hours; wood was added periodically as needed to maintain the desired overall burnrate for the test. Hot start tests were similar to cold start tests with the important difference that the sampling train was not started until the fire was lit and the desired burnrate established.

Emission samples during the Raleigh tests were collected with a modified EPA Method 5 sampling train[8]. For the Boise support tests, emissions were collected with the woodstove dilution sampling system (WSDSS)[9]. Glass fiber filters and XAD-2 adsorbent were used with both trains to collect particles and condensible and semivolatile organics. Teflon coated fiber glass filters (Pallflex) were used with the WSDSS. In all of the tests it was necessary to change the filter several times as the filters became clogged with collected material. Excessive pressure drop was usually encountered within a short time after the addition of fresh fuel to the stove.

Results

The following discussion summarizes the results of bioassay and polynuclear organic material (POM) analyses. Other results, such as total particulate and organic extractable emission rates, have been reported previously[4-7].

PAH emissions - Figures 1 and 2 illustrate the relationship between the PAH emission rates expressed in g/hr and g/kg of wood burned, respectively, and the burnrate. The sum of the specific PAHs quantified, when expressed in g/hr, showed a weak correlation (r^2 = 0.42) with burnrate but no correlation when expressed in g/kg of wood burned. Figure 2, in particular, also shows the effect of wood species, pine producing more PAH than oak. It is important to note that the PAH-burnrate relationship for all the data seems to be direct as compared to the inverse relationship between total train capture emissions and burnrate shown in Figure 3. Total train capture includes the traditional particulates and the semivolatile organics. This indicates that, for these tests, as total emissions

decreased with increasing burnrate, the percent of the emissions constituting the PAH fraction increased. These figures illustrate the effect of stove type with the catalytic stove showing lower emission rates of PAHs and total train capture. Note that, in general, the catalytic stove burning pine produced more PAHs than the conventional stoves burning oak. It would have been interesting to have seen the PAH emission rate for the catalytic stove burning oak, but this test was not included in the study.

An analysis of variance (ANOVA) performed on these data showed few main effects because of the wide variability. One of the main effects identified was the influence of altitude on PAH emissions mentioned previously. The statistical analysis shows that increasing altitude from 90 to 825 m caused a decrease in PAH emission concentration (g/m^3), emission rate (g/hr), and emission factor (g/kg of fuel burned). The statistical analysis confirmed that burnrate exerts a direct effect on PAH emissions (in g/hr only) but not as a main effect (see again Figure 1).

Mutagenicity of emissions - Mutagenicity was determined for each sample using the standard Ames plate incorporation histidine reversion assay. The TA98 strain of bacteria was used, with (+S9) and without (-S9) activation. With activation (+S9) represents the situation where a substance must be metabolized before it is mutagenic. These substances are called indirect acting mutagens. Without activation (-S9) detects substances which are direct acting mutagens.

An analysis of variance was performed separately on the data for the Raleigh and Boise source laboratory tests. The ANOVA results for the Raleigh data are:

- o Increasing burnrate caused mutagenicity (+S9 only) to increase, significant at the 90% confidence interval (CI).
- o Changing from yellow pine to oak caused mutagenicity (both + and - S9) to decrease at just below the 90% CI.
- o Increasing the wood load decreased mutagenicity (-S9 only).

The ANOVA results for the Boise source laboratory tests showed:

- o Changing from the conventional stove to the catalytic stove caused an increase in mutagenicity (both + and - S9) significant at the 99% confidence bound (CB) on the basis of revertants per microgram of sample.
- o Changing from the conventional stove to the catalytic stove caused an increase in mutagenicity (both + and - S9) significant at the 90% CB on the basis of revertants per hour.
- o Changing from western pine to oak caused an increase in mutagenicity (-S9 only) significant at the 95% CB.
- o Changing from western pine to oak caused a decrease in mutagenicity (+S9 only) significant at the 90% CB.

A multiple linear regression analysis was performed on the combined data set. This analysis tended to confirm the ANOVA results on the individual data sets. The following results all relate to a 90% confidence interval:

- o Changing from high to low altitude increased PAH emissions (higher at the low altitude). Mutagenicity (+S9 only) was also higher at low altitude.
- o Changing from conventional to catalytic stove increased the mutagenicity (-S9 only) in terms of rev/ug of sample, rev/m^3 of stack gas, rev/hr and rev/kg wood burned.
- o Changing from oak to pine increased the PAH emissions (see Figures 1 and 2). Direct acting mutagenicity (-S9) was higher for emissions from oak, whereas indirect acting mutagenicity (+S9) was higher for emissions from pine as shown in Figure 4.
- o Increasing burnrate increased PAH emissions in terms of g/hr as shown in Figure 1.

Conclusions

In the Integrated Air Cancer Project, field study emission tests on residential sources such as woodstoves are necessary. However, some variables, such as burnrate, are nearly impossible to measure over short time frames of a few hours without causing a major disruption to the residents. The parallel testing of such residential combustion sources under controlled conditions in a laboratory offers the advantage of allowing measurement of all parameters under simulated field conditions. Together, the field and laboratory data provide the means of adequately characterizing these sources.

Combustion in woodstoves is an inherently variable process because of the nonhomogeniety of the fuel and the batch nature of the fueling procedure. Including cold start in the test protocol adds substantially more variability. Even with proper statistical test program design, this variability makes drawing conclusions difficult.

PAH emissions from a woodstove typical of those used in Boise,ID, were lower at Boise's elevation than at near sea level. There was a corresponding change in mutagenicity (+S9 only). Emissions from the catalytic stove were more mutagenic. Burnrate did not show a statistically significant effect on mutagenicity of the Boise-simulate data; however, it showed a positive effect in the Raleigh simulation. No doubt this discrepancy reflects the fact that different stoves and operating procedures were followed in the two test series.

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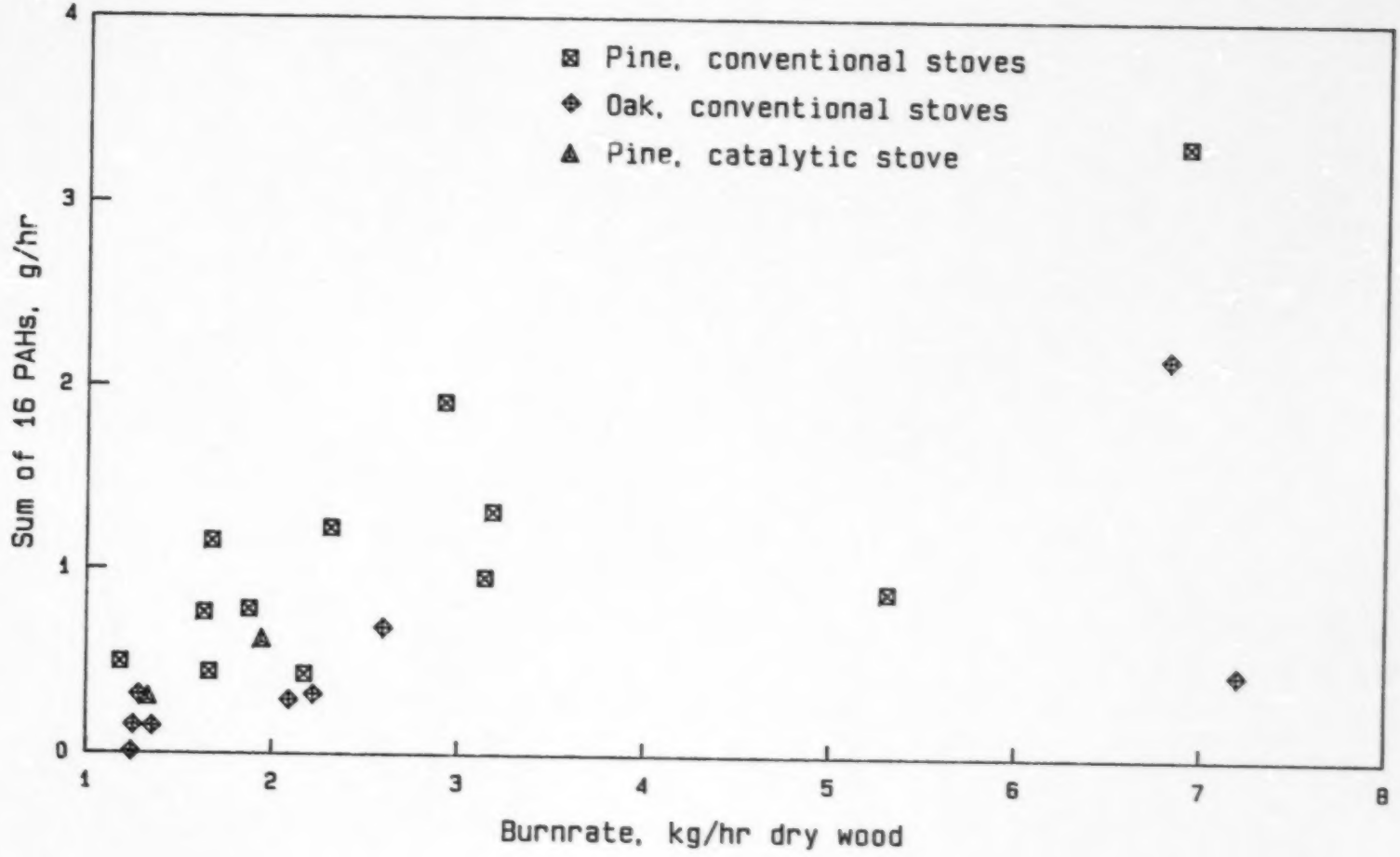

Figure 1. IACP source laboratory woodstove test results for total PAH in g/hr as a function of burnrate, wood species, and stove type.

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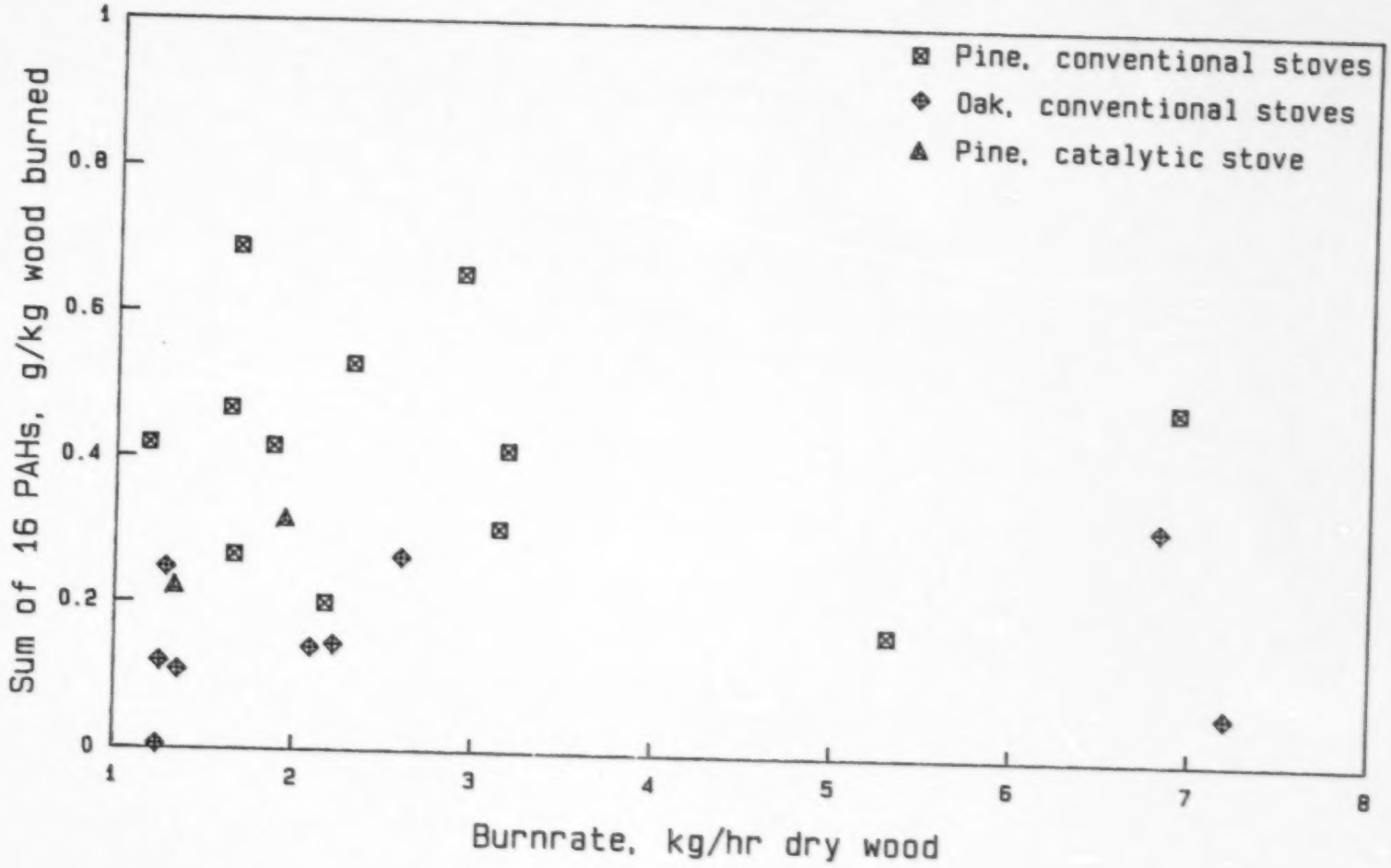

Figure 2. IACP source laboratory woodstove test results for total PAH in g/kg as a function of burnrate, wood species, and stove type.

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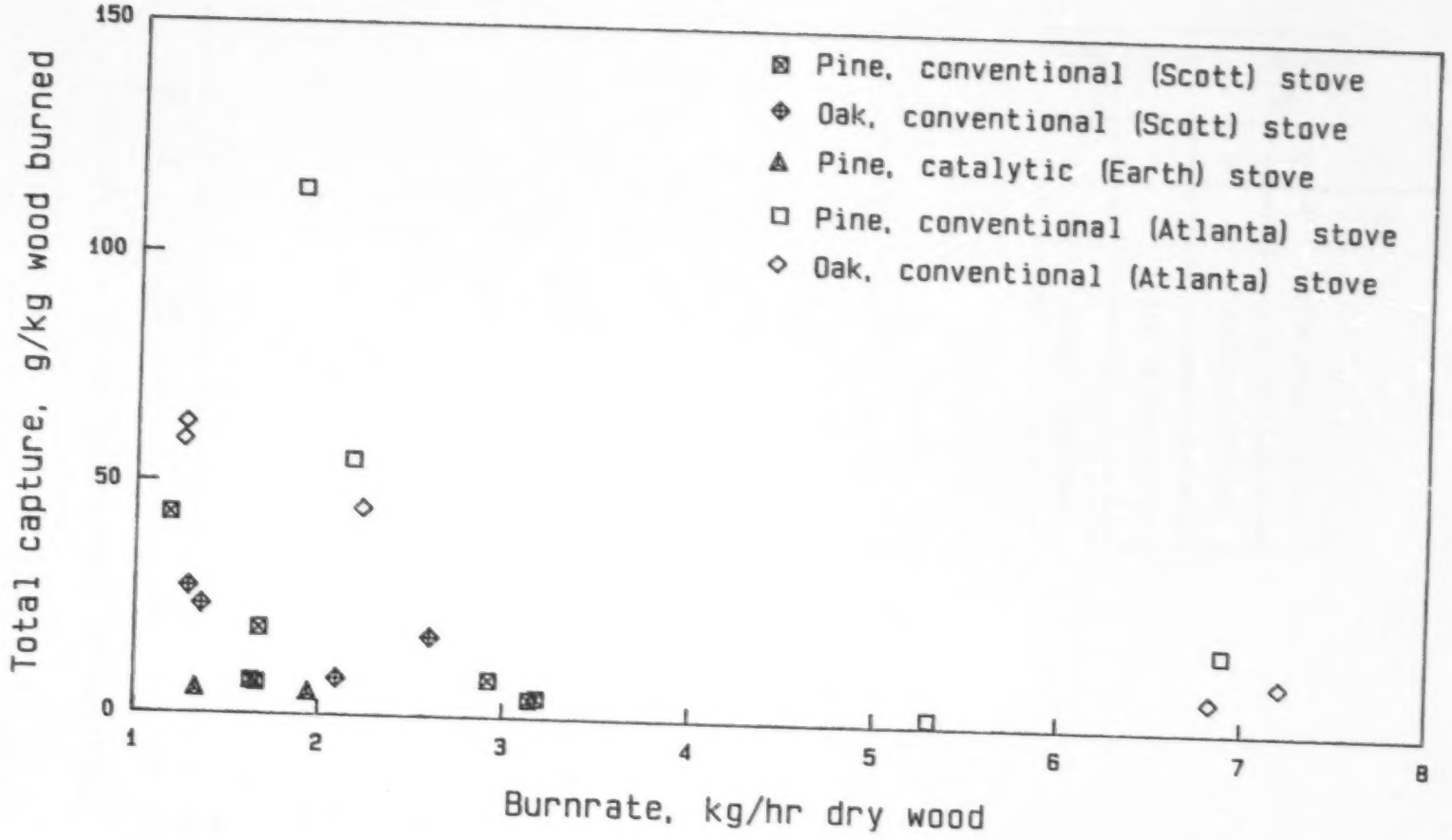

Figure 3. IACP source laboratory woodstove test results for total train catch in g/kg as a function of burnrate, wood species, and stove type.

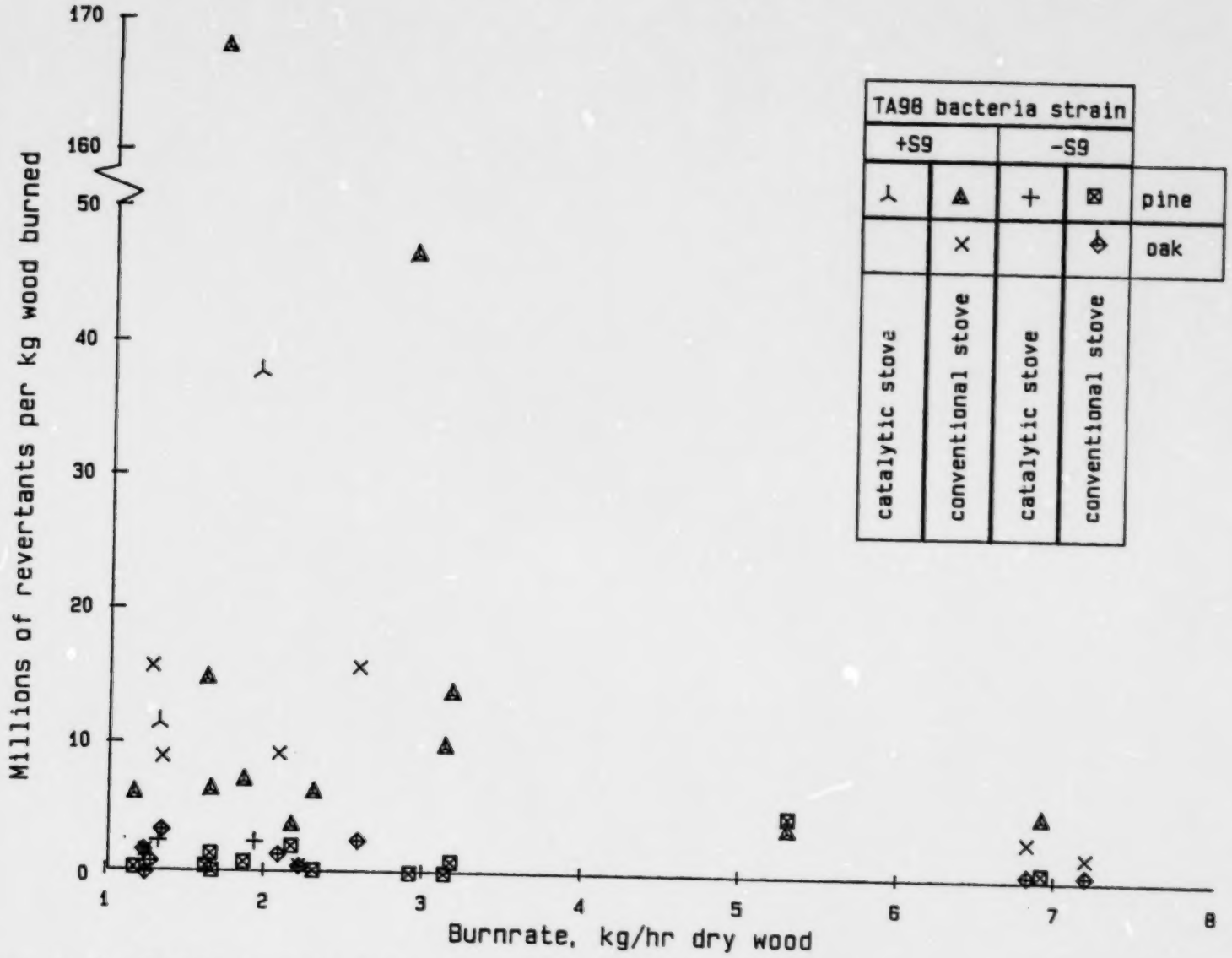

Figure 4. IACP source laboratory woodstove test results for mutagenicity as a function of burnrate, wood species, and stove type.